

# PATENT COOPERATION TREATY

From the  
INTERNATIONAL SEARCHING AUTHORITY

To:

see form PCT/ISA/220

# PCT

## WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1)

Date of mailing  
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference  
see form PCT/ISA/220

**FOR FURTHER ACTION**  
See paragraph 2 below

International application No.  
PCT/CZ2004/000024

International filing date (day/month/year)  
28.04.2004

Priority date (day/month/year)  
30.04.2003

International Patent Classification (IPC) or both national classification and IPC  
A62D3/00

Applicant  
USTAV CHEMICKYCH PROCESU AV CR

### 1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☒ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

### 2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA"). However, this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of three months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

### 3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA:



European Patent Office - P.B. 5818 Patentlaan 2  
NL-2280 HV Rijswijk - Pays Bas  
Tel. +31 70 340 - 2040 Tx: 31 651 epo nl  
Fax: +31 70 340 - 3016

Authorized Officer

Lehnert, A

Telephone No. +31 70 340-4234



**WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING AUTHORITY**

International application No.  
PCT/CZ2004/000024

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**Box No. I    Basis of the opinion**

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1. With regard to the **language**, this opinion has been established on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.
  - ☐ This opinion has been established on the basis of a translation from the original language into the following language , which is the language of a translation furnished for the purposes of international search (under Rules 12.3 and 23.1(b)).
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:
  - a. type of material:
    - ☐ a sequence listing
    - ☐ table(s) related to the sequence listing
  - b. format of material:
    - ☐ in written format
    - ☐ in computer readable form
  - c. time of filing/furnishing:
    - ☐ contained in the international application as filed.
    - ☐ filed together with the international application in computer readable form.
    - ☐ furnished subsequently to this Authority for the purposes of search.
3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

**WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING AUTHORITY**

International application No.  
PCT/CZ2004/000024

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**Box No. II Priority**

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1. ☒ The following document has not been furnished:

☒ copy of the earlier application whose priority has been claimed (Rule 43bis.1 and 66.7(a)).

☐ translation of the earlier application whose priority has been claimed (Rule 43bis.1 and 66.7(b)).

Consequently it has not been possible to consider the validity of the priority claim. This opinion has nevertheless been established on the assumption that the relevant date is the claimed priority date.

2. ☐ This opinion has been established as if no priority had been claimed due to the fact that the priority claim has been found invalid (Rules 43bis.1 and 64.1). Thus for the purposes of this opinion, the international filing date indicated above is considered to be the relevant date.

3. Additional observations, if necessary:

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**Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

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1. Statement

Novelty (N)	Yes: Claims	
	No: Claims	1-3
Inventive step (IS)	Yes: Claims	
	No: Claims	1-3
Industrial applicability (IA)	Yes: Claims	1-3
	No: Claims	

2. Citations and explanations

**see separate sheet**

**WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING  
AUTHORITY (SEPARATE SHEET)**

PCT/CZ2004/000024

**Re Item V.**

- 1 The following documents are referred to in this communication:
  - D1 : US 5 276 250 A (HAGENMAIER HANSPAUL ET AL) 4 January 1994 (1994-01-04)
  - D2 : US 6 303 812 B1 (SAVIDAKIS MICHAEL C ET AL) 16 October 2001 (2001-10-16)
  - D3 : US 3 989 806 A (HYATT DAVID E) 2 November 1976 (1976-11-02)
  - D4 : EP 1 048 327 A (TAGAWA SADAOKO ; SHINETSU CHEMICAL CO (JP)) 2 November 2000 (2000-11-02)
- 2 NOVELTY
  - 2.1 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-3 is not new in the sense of Article 33(2) PCT. Document D1 (see claims, example 17) discloses the dehalogenation of halogenated aromatic compounds at 350 - 500 °C in the presence of a mixed CuO/Cr2O3 catalyst on a silica support. The material to be detoxified may be fly ash. The Cr2O3 has the character of a reducing substance. Metallic copper may be used as well.
  - 2.2 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-3 is not new in the sense of Article 33(2) PCT. Document D2 (see column 2, lines 5 - 18) discloses the dehalogenation of haloaromatic compounds in the presence of copper and propionic acid as a reducing agent.
  - 2.3 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-3 is not new in the sense of Article 33(2) PCT. Document D3 discloses the dehalogenation of hexachlorobenzene with copper on zeolite (reducible copper halides may be used) at temperatures between 300 and 500 °C.
  - 2.4 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claim 1 is not new in the sense of Article 33(2) PCT. Document D4 discloses copper (see paragraph [0024]) in admixture with a

**WRITTEN OPINION OF THE  
INTERNATIONAL SEARCHING  
AUTHORITY (SEPARATE SHEET)**

International application No.

PCT/CZ2004/000024

hydrogen-containing organosilicon compound, used for dehalogenation of  
chlorinated organic compounds (see claims 1-4).

# PATENT COOPERATION TREATY

From the

INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

<b>TRAPLOVA HAKR KUBÁT</b> <b>Law and Patent Offices</b>
<div style="font-size: 1.2em;">14-06-2005</div> <div style="font-size: 1.5em; font-weight: bold; margin-top: 10px;">RECEIVED</div>

## PCT

**WRITTEN OPINION OF THE  
INTERNATIONAL PRELIMINARY  
EXAMINING AUTHORITY**

(PCT Rule 66)

<b>To:</b>  Hakr, Eduard TRAPLOVA HAKR KUBÁT Law and Patent Offices Pristavni 24 170 00 Praha 7 REPUBLIQUE TCHEQUE	<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; padding: 5px;"> <b>Date of mailing</b>  <i>(day/month/year)</i> </td> <td style="width: 50%; padding: 5px;"> <b>01.06.2005</b> </td> </tr> </table>	<b>Date of mailing</b> <i>(day/month/year)</i>	<b>01.06.2005</b>
<b>Date of mailing</b> <i>(day/month/year)</i>	<b>01.06.2005</b>		
<b>Applicant's or agent's file reference</b> 150376/HK	<b>REPLY DUE</b> <b>within 2 month(s)</b> from the above date of mailing		
<b>International application No.</b> PCT/CZ2004/000024	<b>International filing date</b> <i>(day/month/year)</i> 28.04.2004		
<b>Priority date</b> <i>(day/month/year)</i> 30.04.2003			
<b>International Patent Classification (IPC) or both national classification and IPC</b> A62D3/00			
<b>Applicant</b> USTAV CHEMICKYCH PROCESU AV CR ET AL.			

1.	<input checked="" type="checkbox"/> The written opinion established by the International Searching Authority: <div style="margin-left: 20px;"> <input checked="" type="checkbox"/> is      <input type="checkbox"/> is not                      considered to be a written opinion of the International Preliminary Examining Authority                 </div>
2.	This second report contains indications relating to the following items: <div style="margin-left: 20px;"> <input checked="" type="checkbox"/> Box No. I      Basis of the opinion  <input type="checkbox"/> Box No. II      Priority  <input type="checkbox"/> Box No. III      Non-establishment of opinion with regard to novelty, inventive step and industrial applicability  <input type="checkbox"/> Box No. IV      Lack of unity of invention  <input checked="" type="checkbox"/> Box No. V      Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement  <input type="checkbox"/> Box No. VI      Certain documents cited  <input type="checkbox"/> Box No. VII      Certain defects in the international application  <input type="checkbox"/> Box No. VIII      Certain observations on the international application                 </div>
3.	The applicant is hereby invited to reply to this opinion.  <div style="margin-left: 20px;"> <b>When?</b>      See the time limit indicated above. The applicant may, before the expiration of that time limit, request this Authority to grant an extension, see Rule 66.2(e).  <b>How?</b>      By submitting a written reply, accompanied, where appropriate, by amendments, according to Rule 66.3. For the form and the language of the amendments, see Rules 66.8 and 66.9.  <b>Also:</b>      For the examiner's obligation to consider amendments and/or arguments, see Rule 66.4bis. For an informal communication with the examiner, see Rule 66.6. For an additional opportunity to submit amendments, see Rule 66.4.  <b>If no reply is filed,</b> the international preliminary examination report will be established on the basis of this opinion.                 </div>
4.	The final date by which the international preliminary report on patentability (Chapter II of the PCT) must be established according to Rule 69.2 is: 30.08.2005

<b>Name and mailing address of the international preliminary examining authority:</b>  <div style="display: flex; align-items: center;"> <div>                         European Patent Office - P.B. 5818 Patentlaan 2                          NL-2280 HV Rijswijk - Pays Bas                          Tel. +31 70 340 - 2040 Tx: 31 651 epo nl                          Fax: +31 70 340 - 3016                     </div> </div>	<b>Authorized Officer</b>  Lehnert, A  Telephone No. +31 70 340-4234
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**WRITTEN OPINION OF THE INTERNATIONAL  
PRELIMINARY EXAMINING AUTHORITY**

International application No.  
PCT/CZ2004/000024

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**Box No. I Basis of the opinion**

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1. With regard to the **language**, this opinion is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
- ☐ This opinion is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
- ☐ international search (under Rules 12.3 and 23.1(b))
  - ☐ publication of the international application (under Rule 12.4)
  - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements** of the international application, this opinion is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this opinion as "originally filed")*:

**Description, Pages**

1-13 as originally filed

**Claims, Numbers**

1-3 as originally filed

- ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing.
3. ☐ The amendments have resulted in the cancellation of:
- ☐ the description, pages
  - ☐ the claims, Nos.
  - ☐ the drawings, sheets/figs
  - ☐ the sequence listing (*specify*):
  - ☐ any table(s) related to sequence listing (*specify*):
4. ☐ This opinion has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
- ☐ the description, pages
  - ☐ the claims, Nos.
  - ☐ the drawings, sheets/figs
  - ☐ the sequence listing (*specify*):
  - ☐ any table(s) related to sequence listing (*specify*):

**WRITTEN OPINION OF THE INTERNATIONAL  
PRELIMINARY EXAMINING AUTHORITY**

International application No.  
PCT/CZ2004/000024

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**Box No. V Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or  
industrial applicability; citations and explanations supporting such statement**

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**1. Statement**

Novelty (N)	Yes: Claims	
	No: Claims	1-3
Inventive step (IS)	Yes: Claims	
	No: Claims	1-3
Industrial applicability (IA)	Yes: Claims	1-3
	No: Claims	

**2. Citations and explanations:**

**see separate sheet**



**WRITTEN OPINION OF THE INTERNATIONAL  
PRELIMINARY EXAMINING AUTHORITY  
(SEPARATE SHEET)**

International application No.

PCT/CZ2004/000024

**Re Item V.**

- 1 The following documents are referred to in this communication:

D1 : US 5 276 250 A (HAGENMAIER HANSPAUL ET AL) 4 January 1994 (1994-01-04)  
D2 : US 6 303 812 B1 (SAVIDAKIS MICHAEL C ET AL) 16 October 2001 (2001-10-16)  
D3 : US 3 989 806 A (HYATT DAVID E) 2 November 1976 (1976-11-02)  
D4 : EP 1 048 327 A (TAGAWA SADAOKO ; SHINETSU CHEMICAL CO (JP)) 2 November 2000 (2000-11-02)

2 NOVELTY

In response to the applicant's letter of 10.09.2004 the novelty objections based on D1 - D4 are maintained. A document is novelty destroying as long as it discloses (explicitly or implicitly) all the technical features of a claim.

- 2.1 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-3 is not new in the sense of Article 33(2) PCT.  
Document D1 (see claims, example 17) discloses the dehalogenation of halogenated aromatic compounds at 350 - 500 °C in the presence of a mixed CuO/Cr<sub>2</sub>O<sub>3</sub> catalyst on a silica support. The material to be detoxified may be fly ash. The Cr<sub>2</sub>O<sub>3</sub> has the character of a reducing substance. Metallic copper may be used as well.
- 2.2 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-3 is not new in the sense of Article 33(2) PCT.  
Document D2 (see column 2, lines 5 - 18) discloses the dehalogenation of haloaromatic compounds in the presence of copper and propionic acid as a reducing agent.
- 2.3 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-3 is not new in the sense of Article 33(2) PCT.  
Document D3 discloses the dehalogenation of hexachlorobenzene with copper on

**WRITTEN OPINION OF THE INTERNATIONAL  
PRELIMINARY EXAMINING AUTHORITY  
(SEPARATE SHEET)**

International application No.

PCT/CZ2004/000024

zeolite (reducible copper halides may be used) at temperatures between 300 and 500 °C.

2.4 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claim 1 is not new in the sense of Article 33(2) PCT.

Document D4 discloses copper (see paragraph [0024]) in admixture with a hydrogen-containing organosilicon compound, used for dehalogenation of chlorinated organic compounds (see claims 1-4).



**TRAPLOVÁ • HAKR • KUBÁT**  
**Law and Patent Offices**

Patents, Trademarks, Designs, Licences

**10/582649**

**AP3 Rec'd PCT/PTO 13 JUN 2005**

Prague, July 27, 2005

**European Patent Office**  
**P.B. 5818 Patentlaan 2**  
**NL-2280 HV Rijswijk - Pays Bas**

**Re: International Application No. PCT/CZ2004/000024**  
**Applicant: USTAV CHEMICKYCH PROCESU AV CR ET AL.**  
**Priority date: 30.04.2003**  
**International filing date: 28.04.2004**  
  
Your ref.: PCT/CZ2004/000024  
Our ref.: 150376/HK

**Applicant's comments on the Written Opinion of the International Preliminary Examining Authority**

In view of that the Written opinion of the International preliminary examining authority only repeats both novelty and inventiveness objections as already mentioned in the Written opinion of the International searching authority and taking into account that the Applicant's argumentation concerning these objections remained without being counter-argued by the International searching authority at all, the Applicant presently submits the same argumentation as already comprised in his reply to the Written opinion of the International searching authority as his present position to the objections now repeatedly comprised in the Written opinion of the International preliminary examining authority. This argumentation is as comes after:

According to Claim 1 of the present application, the subject-matter of the present application is the method for dehalogenation detoxification of halogenated and/or cyclic compounds wherein at least one halogenated aromatic and/or



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Na Příkopě 20, Prague 1  
Account No. 3483-6151 (USD)  
3400-6151 (EUR)

cyclic compound is heated on a support matrix in a closed system at a temperature of 200 to 500 °C in the presence of copper in metallic form and/or in the form of copper compounds, a hydrogen donor, carbon and at least one additional reducing substance, capable of reducing cupric and cuprous ions to elemental copper at the above temperature.

According to the second complete paragraph of page 5 of the description of the present application "the use of at least one additional reducing substance in addition to carbon, which is capable of reducing cuprous and cupric ions to elemental copper (further only "ARS") at the temperature of the dehalogenation process, constitutes the substance of the invention as, in this case, the reversible process  $\text{Cu} \rightarrow \text{Cu}^{1+} \rightarrow \text{Cu}^{2+}$  and back occurs, in which the nascent form of copper is formed repeatedly and enables the successful course of the dehalogenation process according to the invention. The ability of this nascent form of copper to form an intermediate complex of the compound on the aromatic ring is so high that dehalogenation also occurs in positions that are thermodynamically very stable, so that under optimized conditions, a degree of dehalogenation of up to 99,9%, is attained even for highly stable halogenated aromatic and/or cyclic compounds".

In addition, it ensues from Examples of the present application that the invention dehalogenation method proceeds in the presence of the ARS extremely promptly at relatively low temperatures (in about hours at temperatures less than 300 °C).

The ARS acts during the invention dehalogenation process as a reaction component (it takes part in dehalogenation reaction and just this reaction behavior enables, contrary to catalytic dehalogenation processes, the very prompt course of the dehalogenation) rather than a mere catalyst since it changes its valence state therein.

#### **To the IPEA novelty objection 2.1**

The objection is based on the following: Document D1 (see claims, examples 17) discloses the dehalogenation of halogenated aromatic compounds at 350 - 500 °C in the presence of a mixed  $\text{CuO/Cr}_2\text{O}_3$  catalyst on silica support.

The material to be detoxified may be fly ash. **The  $\text{Cr}_2\text{O}_3$  has the character of a reducing substance.** Metallic copper may be used as well.

The IPEA is incorrect when taking  $\text{Cr}_2\text{O}_3$  for the ARS for the following reasons:

1) The basic knowledge discovered by the present invention was that the presence of the ARS in the system halogenated compound/copper(compound)/hydrogen donor/carbon extremely shortens the dehalogenation times to several hours which is not the case of the same system but containing  $\text{Cr}_2\text{O}_3$ . So for instance in Example 17 of D1, the decomposition of HCB into the extent of 99,9% is reached at the temperature of 450 °C only after 300 hours (compare with 100% dehalogenation of HCB as attained for instance in Example 1 of the present application at the temperature of only 200 °C already after 4 hours when using as the ARS citric acid). From the foregoing, it obviously follows that  $\text{Cr}_2\text{O}_3$  behaves during D1 process as a classic decomposition catalyst (after all, it is so indicated in relevant Examples of D1 and the process itself is there denominated as the decomposition rather than the dehalogenation; the thing is, Examples of D1 are absolutely silent on the products to which the starting halogenated compounds are in fact converted which does not exclude the possibility that the decomposition products are still halogenated and that the complete dehalogenation, as reached by the invention, is not in fact the case of D1) not acting as the invention ARS and thus not converting  $\text{Cu}^{1+}$  and  $\text{Cu}^{2+}$  ions into the nascent form of elemental copper in the mentioned system. Thus it may be concluded that the subject-matter of the present application is novel over D1 process since the latter lacks one of the obligatory process component, i.e. the ARS. This conclusion is also supported by the fact that nowhere in D1 at all a reaction mechanism taking profit from the nascent form of elemental copper is either mentioned or at least suggested.

#### **To the IPEA novelty objection 2.2**

The objection is based on the following: Document D2 (see column 2, lines 5 - 18) discloses the dehalogenation of haloaromatic compounds in the presence of copper and propionic acid as a reducing agent.

The dehalogenation of D2 is different enough from that of the present application that it can not be relevant as novelty objection against subject-matter of the present application. The differences are the following: 1) D2 concerns a partial dehalogenation whereas the complete dehalogenation is reached by the method of the present application; 2) D2 mentions the dehalogenation temperature of from about 100 to about 200 °C, whereas the present application claims the dehalogenation temperature of 200 to 500 °C; 3) D2 lacks a hydrogen donor that is, on the contrary, the obligatory feature of the present patent application; 4) D2 lacks carbon that is, on the contrary, the obligatory feature of the present application; 5) finally, the dehalogenation of D2 is carried out under air atmosphere (at least no mention of that the dehalogenation must proceed in a closed system is given in D2) whereas the dehalogenation according to the present application has to be realized in a closed system ensuring a reducing reaction atmosphere (carbon plus oxygen of the closed system  $\rightarrow$  CO) that is the only possible for causing the dehalogenation to proceed via the mechanism of nascent form of elemental copper. It should therefore be concluded, on the base of the foregoing, that the subject-matter of the present application is novel over D2.

### **To the IPEA novelty objection 2.3**

The objection is based on the following: *Document D3 discloses the dehalogenation of hexachlorobenzene with copper on zeolite (reducible copper halides may be used) at the temperature between 300 and 500 °C.*

In addition to that the dehalogenation of D2 does not include the use of the ARS which is, on the contrary the obligatory feature of the dehalogenation of the present application, the basic difference between the dehalogenation of D3 and that of the present application is that the first of the both dehalogenation is oxidation process whereas the second one must proceed under reducing conditions (see the above note made in connection with the difference 5) of the novelty objection 2.2). The dehalogenation of D3 is thus evidently different from that of the present application which renders the subject-matter of the present application novel over D3.

**To the IPEA novelty objection 2.4**

The objection is based on the following: Document D4 discloses copper (see paragraph [0024] in admixture with a hydrogen-containing organosilicon compound, used for dehalogenation of chlorinated organic compounds (see claims 1-4).

At the first sight, the dehalogenation of D4 considerably differs from that of the present application. The main difference between the both dehalogenations consists in that the dehalogenation of D4 uses an organosilicon compound having a hydrogen atom directly attached to a silicon atom (that is not, on the contrary, included in the dehalogenation of the present patent application) and takes profit from high reactivity of said hydrogen atom (on contrary, the dehalogenation process of the patent application is based on high reactivity of the nascent form of elemental copper). From this, further differences derive: the dehalogenation of D4 does not include the ARS and is not carried out in a closed system these two measures being, on the contrary, the obligatory features of the dehalogenation process according to the present application. Taking into account the foregoing the subject-matter of the present application therefore appears to be incontestably novel over D4.

In behalf of  
USTAV CHEMICKYCH PROCESU AV CR ET AL.

Eduard Hakr   
European Patent Attorney



**TRAPLOVÁ • HAKR • KUBÁT**  
**Law and Patent Offices**

Patents, Trademarks, Designs, Licences

Prague, September 10, 2004

**International Bureau of WIPO,  
34 chemin des Colombettes  
1211 Geneva 20,  
Switzerland**

**Re: International Application No. PCT/CZ2004/000024  
Applicant: USTAV CHEMICKYCH PROCESU AV CR  
Priority date: 30.04.2003  
International filing date: 28.04.2004**

Your ref.: PCT/CZ2004/000024

Our ref.: 150376/HK

**Comments on the Written Opinion of the International  
Searching Authority**

According to Claim 1 of the present application, the subject-matter of the present application is the method for dehalogenation detoxification of halogenated and/or cyclic compounds wherein at least one halogenated aromatic and/or cyclic compound is heated on a support matrix in a closed system at a temperature of 200 to 500 °C in the presence of copper in metallic form and/or in the form of copper compounds, a hydrogen donor, carbon and at least one additional reducing substance, capable of reducing cupric and cuprous ions to elemental copper at the above temperature.

According to the second complete paragraph of page 5 of the description of the present application "the use of at least one additional reducing substance in addition to carbon, which is capable of reducing cuprous and cupric ions to elemental copper (further only "ARS") at the temperature of



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E-mail: thk@thk.cz  
www.thk.cz

Bank: Živnostenská banka  
Na Příkopě 20, Prague 1  
Account No. 3483-6151 (USD)  
3400-6151 (EUR)



the dehalogenation process, **constitutes the substance of the invention** as, in this case, the reversible process  $\text{Cu} \rightarrow \text{Cu}^{1+} \rightarrow \text{Cu}^{2+}$  and back occurs, in which **the nascent form of copper** is formed repeatedly and enables the successful course of the dehalogenation process according to the invention. The ability of this nascent form of copper to form an intermediate complex of the compound on the aromatic ring is so high that dehalogenation also occurs in positions that are thermodynamically very stable, so that under optimized conditions, a degree of dehalogenation of up to 99,9%, is attained even for highly stable halogenated aromatic and/or cyclic compounds".

In addition, it ensues from Examples of the present application that the invention dehalogenation method proceeds in the presence of the ARS extremely promptly at relatively low temperatures (in about hours at temperatures less than 300 ° C).

The ARS acts during the invention dehalogenation process as a reaction component (it takes part in dehalogenation reaction and just this reaction behavior enables, contrary to catalytic dehalogenation processes, the very prompt course of the dehalogenation) rather than a mere catalyst since it changes its valence state therein.

#### **To the ISA novelty objection 2.1**

The objection is based on the following: Document D1 (see claims, examples 17) discloses the dehalogenation of halogenated aromatic compounds at 350 - 500 °C in the presence of a mixed  $\text{CuO/Cr}_2\text{O}_3$  catalyst on silica support. The material to be detoxified may be fly ash. **The  $\text{Cr}_2\text{O}_3$  has the character of a reducing substance.** Metallic copper may be used as well.

The ISA is incorrect when taking  $\text{Cr}_2\text{O}_3$  for the ARS for the following reasons:

- 1) The basic knowledge discovered by the present invention was that the presence of the ARS in the system halogenated compound/copper(compound)/hydrogen donor/carbon extremely shortens the dehalogenation times to several hours which is not the case of the same system but containing  $\text{Cr}_2\text{O}_3$ . So for instance in Example 17 of D1, the decomposition of HCB into the extent of 99,9% is reached at the temperature of

450 °C only after 300 hours (compare with 100% dehalogenation of HCB as attained for instance in Example 1 of the present application at the temperature of only 200 °C already after 4 hours when using as the ARS citric acid). From the foregoing, it obviously follows that  $\text{Cr}_2\text{O}_3$  behaves during D1 process as a classic decomposition catalyst (after all, it is so indicated in relevant Examples of D1 and the process itself is there denominated as the decomposition rather than the dehalogenation; the thing is, Examples of D1 are absolutely silent on the products to which the starting halogenated compounds are in fact converted which does not exclude the possibility that the decomposition products are still halogenated and that the complete dehalogenation, as reached by the invention, is not in fact the case of D1) not acting as the invention ARS and thus not converting  $\text{Cu}^{1+}$  and  $\text{Cu}^{2+}$  ions into the nascent form of elemental copper in the mentioned system. Thus it may be concluded that the subject-matter of the present application is novel over D1 process since the latter lacks one of the obligatory process component, i.e. the ARS. This conclusion is also supported by the fact that nowhere in D1 at all a reaction mechanism taking profit from the nascent form of elemental copper is either mentioned or at least suggested.

#### **To the ISA novelty objection 2.2**

The objection is based on the following: Document D2 (see column 2, lines 5 - 18) discloses the dehalogenation of haloaromatic compounds in the presence of copper and propionic acid as a reducing agent.

The dehalogenation of D2 is different enough from that of the present application that it can not be relevant as novelty objection against subject-matter of the present application. The differences are the following: 1) D2 concerns a partial dehalogenation whereas the complete dehalogenation is reached by the method of the present application; 2) D2 mentions the dehalogenation temperature of from about 100 to about 200 °C, whereas the present application claims the dehalogenation temperature of 200 to 500 °C; 3) D2 lacks a hydrogen donor that is, on the contrary, the obligatory feature of the present patent application; 4) D2 lacks carbon that is, on the contrary,

the obligatory feature of the present application; 5) finally, the dehalogenation of D2 is carried out under air atmosphere (at least no mention of that the dehalogenation must proceed in a closed system is given in D2) whereas the dehalogenation according to the present application has to be realized in a closed system ensuring a reducing reaction atmosphere (carbon plus oxygen of the closed system  $\rightarrow$  CO) that is the only possible for causing the dehalogenation to proceed via the mechanism of nascent form of elemental copper. It should therefore be concluded, on the base of the foregoing, that the subject-matter of the present application is novel over D2.

#### **To the ISA novelty objection 2.3**

The objection is based on the following: *Document D3 discloses the dehalogenation of hexachlorobenzene with copper on zeolite (reducible copper halides may be used) at the temperature between 300 and 500 °C.*

In addition to that the dehalogenation of D2 does not include the use of the ARS which is, on the contrary the obligatory feature of the dehalogenation of the present application, the basic difference between the dehalogenation of D3 and that of the present application is that the first of the both dehalogenation is oxidation process whereas the second one must proceed under reducing conditions (see the above note made in connection with the difference 5) of the novelty objection 2.2). The dehalogenation of D3 is thus evidently different from that of the present application which renders the subject-matter of the present application novel over D3.

#### **To the ISA novelty objection 2.4**

The objection is based on the following: *Document D4 discloses copper (see paragraph [0024] in admixture with a hydrogen-containing organosilicon compound, used for dehalogenation of chlorinated organic compounds (see claims 1-4).*

At the first sight, the dehalogenation of D4 considerably differs from that of the present application. The main difference between the both dehalogenations consists in that the dehalogenation of D4 uses an organosilicon compound having a hydrogen atom directly attached to a

silicon atom (that is not, on the contrary, included in the dehalogenation of the present patent application) and takes profit from high reactivity of said hydrogen atom (on contrary, the dehalogenation process of the patent application is based on high reactivity of the nascent form of elemental copper). From this, further differences derive: the dehalogenation of D4 does not include the ARS and is not carried out in a closed system these two measures being, on the contrary, the obligatory features of the dehalogenation process according to the present application. Taking into account the foregoing the subject-matter of the present application therefore appears to be incontestably novel over D4.

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